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**BOX: PATENT APPLICATION**

Assistant Commissioner for Patents  
Washington, D.C. 20231

Re: Application of Kenji MATSUO, Masahiro HOJO, Tomohisa NISHIKAWA and Kazuomi KOBAYASHI  
**RUBBER COMPOSITION AND PNEUMATIC TIRE USING SAID RUBBER COMPOSITION**  
Our Reference: Q54675

Dear Sir:

Attached hereto is the application identified above including the specification, claims, executed Declaration and Power of Attorney, two (2) sheets of drawings, one (1) priority document, executed Assignment and PTO Form 1595.

The Government filing fee is calculated as follows:

Total Claims	20 - 20 =	0 x \$18 =	\$ 000.00
Independent Claims	3 - 3 =	0 x \$78 =	\$ 000.00
Base Filing Fee	(\$760.00)		\$ 760.00
Multiple Dep. Claim Fee	(\$260.00)		\$ 000.00
<b>TOTAL FILING FEE</b>			<b>\$ 760.00</b>
Recordation of Assignment Fee			\$ 40.00
<b>TOTAL U.S. GOVERNMENT FEE</b>			<b>\$ 800.00</b>

Checks for the statutory filing fee of \$ 760.00 and Assignment recordation fee of \$ 40.00 are attached. You are also directed and authorized to charge or credit any difference or overpayment to Deposit Account No. 19-4880. The Commissioner is hereby authorized to charge any fees under 37 C.F.R. 1.16 and 1.17 and any petitions for extension of time under 37 C.F.R. 1.136 which may be required during the entire pendency of the application to Deposit Account No. 19-4880. A duplicate copy of this transmittal letter is attached.

Priority is claimed from:

Japanese Patent Application

10-159512

Filing Date

June 8, 1998

The Office is invited to contact the above firm on any question which might arise on the above-named application. Any contact that the Office might need to make should be directed to the undersigned at (202)293-7060.

Respectfully submitted,  
SUGHRUE, MION, ZINN, MACPEAK & SEAS  
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By

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# RUBBER COMPOSITION AND PNEUMATIC TIRE USING SAID RUBBER COMPOSITION

## BACKGROUND OF THE INVENTION

### Field of the Invention

The present invention relates to a rubber composition and a pneumatic tire and, more particularly, to a rubber composition having improved heat resistance and a pneumatic tire using said rubber composition.

### Description of the Related Art

Heretofore, a reinforcing layer composed of a rubber composition alone or a composite of a rubber composition and another material such as a fiber is disposed at side wall portions of a pneumatic tire to enhance the rigidity of the side wall portions. However, when the temperature of the rubber composition increases to 200°C or higher, for example, in use of the pneumatic tire in a condition in which the pressure inside the pneumatic tire (hereinafter referred to as the inner pressure) is reduced by a puncture or the like, i.e., in a run-flat condition, the rubber composition tends to show rupture of crosslinked portions obtained by vulcanization or of the polymer itself forming the rubber component. Due to an increase in deformation of the caused by a decrease in modulus side wall portion, heat generation of the rubber composition is accelerated and/or the limit of failure of the rubber lowers. As the result, the tire is damaged in a relatively short time.

To increase the time before the tire is damaged to as long as possible, the modulus of the rubber composition may be increased as much as

possible by modifying the formulation of the rubber composition or the heat generation in the rubber composition itself may be suppressed by decreasing  $\tan \delta$  of the rubber composition as much as possible by modifying the formulation of the rubber composition. However, improvement by modifying the formulation has limit. Further, when the amounts of rubber reinforcing layers and/or bead fillers are increased to ensure durability of a desired level or higher, ride comfort under normal running condition and noise level deteriorate and the weight of the tire increases.

#### SUMMARY OF THE INVENTION

An object of the present invention is to provide a rubber composition having improved heat resistance and a pneumatic tire produced using this rubber composition, more particularly, a pneumatic tire exhibiting improved durability using this rubber composition as a rubber composition for side wall reinforcing layer and/or a rubber composition for bead portion reinforcing rubber filler

As the result of intensive studies by the present inventors on various chemicals used in formulations to improve heat resistance of the rubber composition, it was found that the heat resistance of a rubber composition can be remarkably improved when specific compounds are used in the formulation. The present invention has been completed on the basis of this knowledge.

The object of the present invention can be achieved in accordance with (1) to (18) described in the following.

- (1) A rubber composition having, in a curve exhibiting a change in

dynamic storage modulus during elevation of temperature, an intersection of an extrapolation line A of a portion in which the dynamic storage modulus shows an approximately linear change before a rapid decrease at temperatures higher than 100°C and an extrapolation line B of a portion in which the dynamic storage modulus rapidly decreases, at a temperature of 170°C or higher.

(2) A rubber composition described in (1), which comprises sodium 1,6-hexamethylenedithiosulfate dihydrate.

(3) A rubber composition described in any of (1) and (2), wherein the amount of sodium 1,6-hexamethylenedithiosulfate dihydrate is 1 to 10 parts by weight per 100 parts by weight of a rubber component.

(4) A rubber composition described in any of (1) to (3), which comprises a compound A having two or more ester groups in one molecule.

(5) A rubber composition having, in a curve exhibiting a change in dynamic storage modulus during elevation of temperature, a difference  $\Delta E'$  between the maximum value and the minimum value of the dynamic storage modulus at a temperature between 180 and 200°C of 2.5 MPa or less.

(6) A rubber composition described in (5), which comprises a compound A having two or more ester groups in one molecule.

(7) A rubber composition described in any of (4) and (6), wherein the compound A is an acrylate or a methacrylate.

(8) A rubber composition described in any of (4), (6) and (7), wherein the compound A is a polyfunctional ester of a polyhydric alcohol and acrylic acid or methacrylic acid.

(9) A rubber composition described in any of (4), and (6) to (8),

wherein the polyhydric alcohol forming the compound A is at least one compound selected from the group consisting of tetramethylolmethane, trimethylolpropane and polymers of these compounds.

(10) A rubber composition described in any of (4), and (6) to (9), wherein the polyhydric alcohol is trimethylolpropane or a dimer of tetramethylolmethane.

(11) A rubber composition described in any of (4), and (6) to (10), wherein the amount of the compound A is 0.5 to 20 parts by weight per 100 parts by weight of a rubber component.

(12) A pneumatic tire comprising side wall portions reinforced with a rubber reinforcing layer, wherein a rubber composition for the rubber reinforcing layer comprises the rubber composition described in any of (1) to (11).

(13) A pneumatic tire comprising bead fillers for which the rubber composition described in any of (1) to (11) is used.

(14) A pneumatic tire described in any of (12) and (13), which is a run-flat tire.

(15) A rubber composition for side reinforcing layers and/or bead fillers comprising sodium 1,6-hexamethylenedithiosulfate dihydrate.

(16) A rubber composition described in (15), which comprises a compound A having two or more ester groups in one molecule.

(17) A rubber composition for side reinforcing layers and/or bead fillers which comprises a compound A having two or more ester groups in one molecule.

(18) Use of sodium 1,6-hexamethylenedithiosulfate dihydrate in side reinforcing layers and/or bead fillers of a pneumatic tire.

## BRIEF DESCRIPTION OF THE DRAWINGS

Figure 1 shows a cross-sectional view of an example of the pneumatic tire of the present invention.

Figure 2 shows a curve exhibiting the change in dynamic storage modulus during elevation of temperature.

Figure 3 shows a diagram exhibiting, in the curve exhibiting a change in dynamic storage modulus during elevation of temperature, temperature C at an intersection of extrapolation line A of a portion in which the dynamic storage modulus shows an approximately linear change before rapidly decreasing at temperatures higher than 100°C and extrapolation line B of a portion in which the dynamic storage modulus rapidly decreases.

## DESCRIPTION OF THE PREFERRED EMBODIMENTS

The rubber composition has, in a curve exhibiting the change in dynamic storage modulus  $E'$  during elevation of temperature, the intersection of extrapolation line A of a portion in which the dynamic storage modulus shows an approximately linear change before a rapid decrease at temperatures higher than 100°C and extrapolation line B of a portion in which the dynamic storage modulus rapidly decreases, at a temperature of 170°C or higher.

In the curve exhibiting the change in dynamic storage modulus  $E'$  during elevation of temperature, the temperature at the intersection of extrapolation line A of a portion in which the dynamic storage modulus shows an approximately linear change before a rapid decrease at

temperatures higher than 100°C and extrapolation line B of a portion in which the dynamic storage modulus rapidly decreases is temperature C shown in Figure 3.

When the temperature at the intersection is lower than 170°C, durability of the rubber composition at the high temperatures caused by heat generated in the run-flat condition is not sufficient. Therefore, the temperature must be 170°C or higher.

Extrapolation line A is a line obtained by extrapolation of the portion in which the dynamic modulus shows an approximately linear change before a rapid decrease at temperatures higher than 100°C in the curve exhibiting the change in dynamic storage modulus during elevation of temperature. It is preferable that extrapolation line A contacts the curve showing the change in dynamic storage modulus in a range of 40°C and more preferably in a range of 20°C.

Extrapolation line B is a line obtained by extrapolation of the portion in which the dynamic modulus rapidly decreases. It is preferable that extrapolation line B contacts the curve showing the change in dynamic storage modulus in a range of 15°C and more preferably in a range of 10°C.

In the present invention, it is preferable that sodium 1,6-hexamethylenedithiosulfate dihydrate is used as an agent to prevent heat aging. Sodium 1,6-hexamethylenedithiosulfate dihydrate suppresses rupture of chains of the polymer constituting the rubber component. Therefore, in the curve exhibiting the change in dynamic storage modulus during elevation of temperature, the intersection of the extrapolation line A of the portion in which the dynamic modulus shows an approximately

linear change before a rapid decrease at temperatures higher than 100°C and the extrapolation line B of the portion in which the dynamic modulus rapidly decreases can be easily brought to a temperature of 170°C or higher.

The amount of sodium 1,6-hexamethylenedithiosulfate dihydrate is not particularly limited. From the standpoint of achieving the object of the present invention, it is preferable that the amount is in the range of 1 to 10 parts by weight per 100 parts by weight of the rubber component.

In the present invention, the rubber composition may further comprise a compound having two or more ester groups in one molecule as an agent for preventing degradation.

The agent for preventing degradation preferably used in the present invention is substantially inert during vulcanization at a temperature lower than 170°C. Therefore, the agent for preventing degradation does not take part in the crosslinking at the temperature of vulcanization (generally about 160°C) and modulus does not increase to a value higher than the prescribed value. When the temperature of the rubber composition is 170°C or higher, degradation of the rubber begins to take place and rupture of crosslinking points and/or polymer chains occurs. However, re-crosslinking of the polymer chains with the agent for preventing degradation proceeds at the same time and the decrease in modulus can be suppressed. As the result, heat generation can be suppressed even at high temperatures.

The compound having two or more ester groups in one molecule is not particularly limited. An acrylate or a methacrylate is preferable and a polyfunctional ester formed from a polyhydric alcohol and acrylic acid or



methacrylic acid is more preferable.

Examples of the polyhydric alcohol include alkylene glycols such as methylene glycol, ethylene glycol, propylene glycol, butylene glycol, pentanediol and hexanediol; polymers of alkylene glycols; compounds obtained by introducing methylol groups into the above mentioned alkylene glycols and polymers of alkylene glycols; ketoroses such as erythritol; compounds having polyalkylene oxide groups such as polyalkoxyphenylpropanes; and polyesters and oligoesters having two or more alcoholic hydroxyl groups. Among these compounds, compounds obtained by introducing methylol groups into the above mentioned alkylene glycols and polymers thereof are preferable.

Specific examples of the compounds having two or more ester groups in one molecule include 1,3-butylene glycol diacrylate, 1,5-pentanediol diacrylate, neopentyl glycol diacrylate, 1,6-hexanediol diacrylate, diethylene glycol diacrylate, triethylene glycol diacrylate, tetraethylene glycol diacrylate, polyethylene glycol diacrylate, polypropylene glycol diacrylate, bis(4-acryloxy)polyethoxyphenylpropane oligoester diacrylate, pentaerythritol triacrylate, trimethylolpropane triacrylate, tetramethylolmethane tetraacrylate, dipentaerythritol hexaacrylate, dipentaerythritol pentaacrylate, oligoester polyacrylate, dipropylene glycol dimethacrylate, trimethylolethane trimethacrylate, trimethylolpropane trimethacrylate, di(tetramethylolmethane) pentamethacrylate and di(tetramethylolmethane) trimethacrylate. Among these compounds, di(tetramethylolmethane) pentamethacrylate, di(tetramethylolmethane) trimethacrylate and trimethylolpropane trimethacrylate are preferable. A single type or a mixture of two or more types of the above compounds may be used.

The amount of the compound having two or more ester groups in one molecule is preferably 0.5 to 20 parts by weight and more preferably 1.0 to 15 parts by weight per 100 parts by weight of the rubber component.

In the rubber composition of the present invention, rupture of polymer chains can be suppressed by the agent for preventing heat aging. Even when rupture of polymer chains takes place, the broken polymer chains are reconstructed by the agent for preventing degradation and the change in dynamic storage modulus of the rubber composition in the temperature range of 170 to 200°C can be suppressed to 3 MPa or less. In particular, the rubber composition comprising the agent for preventing degradation of the present invention preferably have a difference  $\Delta E'$  between the maximum value and the minimum value of the dynamic storage modulus at a temperature between 180 and 200°C of 2.5 MPa or less.

The rubber component used in the present invention is not particularly limited and can be suitably selected from generally used rubber components. Examples of the rubber component include natural rubber (NR), synthetic polyisoprene (IR), polybutadiene (BR), styrene-butadiene copolymers (SBR), butyl rubber (IIR) and ethylene-propylene-diene copolymers (EPDM). A single type or a blend of two or more types of the above rubber components may be used.

The rubber composition of the present invention may further comprise suitable compounding ingredients generally used in the rubber industry such as crosslinking agents, crosslinking accelerators, antioxidants, softeners, reinforcing fillers and inorganic fillers. The rubber composition of the present invention may also be used as a

composite together with particles, fibers, cloths and the like made of various materials.

In the pneumatic tire of the present invention, bead fillers and/or rubber reinforcing layers disposed in the side wall portions may preferably comprise the above rubber composition.

The pneumatic tire, particularly the bead filler and the side wall portion of the pneumatic tire, will be briefly described with reference to Figure 1.

Figure 1 shows a cross-sectional view of an example of the pneumatic tire. A carcass 2 has a turned-up carcass ply 2a and a down carcass ply 2b, which have reinforcing cords, disposed in the direction approximately perpendicular to a plane 10. Two layers of belts 3 (steel belts) are disposed on the carcass plies in the radial direction of the tire. A tread rubber 8 is disposed on the belts 3 at the surface portion of the tire which contacts road surfaces. Side wall portions 6 are disposed at both sides of the tread rubber 8 on the carcass ply 2.

Both end portions of the turned-up carcass ply 2 are turned-up around bead cores 4 to form turned-up end portions. Bead fillers 5 made of a hard rubber are disposed between the carcass ply 2 and the turned-up end portions thereof at the outside of the bead cores 4 in the radial direction of the tire. The down carcass ply 2b is disposed extending between the side wall portions 6 and the turned-up end portions of the turned-up carcass ply 2a. Rubber reinforcing layers 7 having a crescent-shaped cross section are disposed at the inner circumferential surface of the side walls portion of the turned-up carcass ply 2a.

It is preferable that at least one of the rubber composition for the

bead filler and for the rubber reinforcing layer comprises sodium 1,6-hexamethylenedithiosulfate dihydrate. It is more preferable that the rubber composition for the bead filler further comprises a compound having two or more ester groups in one molecule. The rubber composition for the bead filler and/or the rubber reinforcing layer preferably has a dynamic storage modulus  $E'$  of 8 MPa or more at 160°C, more preferably 10 MPa or more at 160°C and most preferably 13 MPa or more at 160°C.

The best tire durability may be obtained when both of the rubber members comprise the above described rubber composition.

When conventional pneumatic tires are used in a condition of a markedly reduced inner pressure (in the so-called run-flat condition), deformation of the tire caused by the load markedly increases. Heat generation caused by the deformation increases particularly markedly at side wall portions and damage occurs in the side wall portions. This problem of conventional tires can be overcome by the pneumatic tire comprising the above rubber composition.

Thus, durability of the tire side wall portions can be particularly improved by using the compound described in the present invention in the rubber compositions for the tire and preferably in the rubber compositions for the rubber reinforcing layer of the side wall portions and/or the bead fillers. As the result, for example, the driving distance in the run-flat condition can be remarkably increased. In other words, the present invention can be effectively applied to the run-flat tire which particularly emphasizes safety in the run-flat condition.

## EXAMPLES

The present invention will be described specifically with reference to examples in the following. However, the present invention is not limited by the examples.

In the examples, part and % mean part by weight and % by weight unless otherwise mentioned.

Various measurements were made in accordance with the following methods.

(1) Viscoelastic characteristics of a rubber composition

A slab sheet having a thickness of 2 mm was prepared by vulcanization at 160°C for 12 minutes and a sample having a width of 5 mm and a length of 20 mm was cut out from the slab sheet. Dynamic storage modulus ( $E'$ ) of the sample was measured using SPECTROMETER manufactured by TOYO SEIKI Co., Ltd. at an initial load of 160 g under a dynamic strain of 1% at a frequency of 52 Hz while the temperature was raised from 20 to 250°C at a rate of 3°C/second.

(2) Durability of a tire in the run-flat condition

A tire inflated with an inner pressure of 3.0 kg/cm<sup>2</sup> was fitted to a rim. After the tire was left standing at a room temperature of 38°C for 24 hours, the pressure inside the tire was set to the atmospheric pressure by removing the valve core. Then, the tire was subjected to the drum test under a load of 570 kg at a speed of 89 km/hour at a temperature of 38°C. The distance driven before trouble occurred was taken as durability in the run-flat condition. Durabilities in the run-flat condition in the Examples are expressed as indices relative to the durability of the control tire in Comparative Example 1 which is set at the value of 100. The greater the index, the better the durability in the run-flat condition.

Examples 1-18:

Rubber compositions were prepared in accordance with the formulations shown in Table 1 and the dynamic storage modulus  $E'$  was measured with an elevation of temperature. The results are shown in Table 1.

Table 1 - 1

Example		1	2	3	4	5	6
Comparative Example	1						
Natural rubber	30.0	30.0	30.0	30.0	30.0	30.0	30.0
Butadiene rubber *1	70.0	70.0	70.0	70.0	70.0	70.0	70.0
Carbon black *2	60.0	60.0	60.0	60.0	60.0	60.0	70.0
Softener *3	5.0	5.0	5.0	5.0	5.0	5.0	5.0
Zinc oxide	3.0	3.0	3.0	3.0	3.0	3.0	3.0
Stearic acid	1.0	1.0	1.0	1.0	1.0	1.0	1.0
Antioxidant *4	2.0	2.0	2.0	2.0	2.0	2.0	2.0
Vulcanization accelerator *5	3.5	3.5	3.5	3.5	3.5	3.5	3.5
Agent for preventing heat aging *6	0.0	1.0	2.0	3.0	5.0	10.0	3.0
Agent for preventing degradation *7	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Sulfur	5.0	5.0	5.0	5.0	5.0	5.0	5.0
Temperature C (°C)	169	174	176	178	180	181	178
$\Delta E'$ *8 (MPa)	3.0	3.0	3.0	3.0	3.0	3.0	3.0
Applied portion							
reinforcing rubber	applied	applied	applied	applied	applied	applied	applied
bead filler	not applied	not applied	not applied	not applied	not applied	not applied	not applied
Durability in the run-flat condition	100	105	110	123	132	140	125

Table 1 - 2

Example	7	8	9	10	11	12
Natural rubber	30.0	30.0	30.0	50.0	70.0	30.0
Butadiene rubber <sup>*1</sup>	70.0	70.0	70.0	50.0	30.0	70.0
Carbon black <sup>*2</sup>	50.0	40.0	30.0	60.0	60.0	60.0
Softener <sup>*3</sup>	5.0	5.0	5.0	5.0	5.0	5.0
Zinc oxide	3.0	3.0	3.0	3.0	3.0	3.0
Stearic acid	1.0	1.0	1.0	1.0	1.0	1.0
Antioxidant <sup>*4</sup>	2.0	2.0	2.0	2.0	2.0	2.0
Vulcanization accelerator <sup>*5</sup>	3.5	3.5	3.5	3.5	3.5	3.5
Agent for preventing heat aging <sup>*6</sup>	3.0	3.0	3.0	3.0	3.0	1.0
Agent for preventing degradation <sup>*7</sup>	0.0	0.0	0.0	0.0	0.0	5.0
Sulfur	5.0	5.0	5.0	5.0	5.0	5.0
Temperature C (°C)	178	178	178	178	178	174
$\Delta E'$ <sup>*8</sup> (MPa)	3.0	3.0	3.0	3.0	3.0	2.5
Applied portion						
reinforcing rubber	applied	applied	applied	applied	applied	applied
bead filler	not applied	not applied	not applied	not applied	not applied	not applied
Durability in the run-flat condition	121	119	115	121	119	130



Table 1 - 3

Example	13	14	15	16	17	18
Natural rubber	30.0	30.0	30.0	30.0	30.0	30.0
Butadiene rubber *1	70.0	70.0	70.0	70.0	70.0	70.0
Carbon black *2	60.0	60.0	60.0	60.0	60.0	60.0
Softener *3	5.0	5.0	5.0	5.0	5.0	5.0
Zinc oxide	3.0	3.0	3.0	3.0	3.0	3.0
Stearic acid	1.0	1.0	1.0	1.0	1.0	1.0
Antioxidant *4	2.0	2.0	2.0	2.0	2.0	2.0
Vulcanization accelerator *5	3.5	3.5	3.5	3.5	3.5	3.5
Agent for preventing heat aging *6	2.0	3.0	5.0	10.0	3.0	5.0
Agent for preventing degradation *7	5.0	5.0	5.0	5.0	0.0	0.0
Sulfur	5.0	5.0	5.0	5.0	5.0	5.0
Temperature C (°C)	175	178	180	180	178	178
$\Delta E'$ *8 (MPa)	2.5	2.5	2.5	2.5	3.0	3.0
Applied portion						
reinforcing rubber	applied	applied	applied	applied	applied	applied
bead filler	not applied	not applied	not applied	not applied	applied	applied
Durability in the run-flat condition	134	142	154	166	143	150

The amounts in the above table are all in parts by weight.

\*1: BR01 (a trade mark; manufactured by JSR Corporation)

\*2: FEF

\*3: Spindle oil

\*4: NOCRAC 6C (a trade mark; manufactured by OUCHI SHINKO KAGAKU KOGYO Co., Ltd.)

\*5: NOCCOLOR NS (a trade mark; manufactured by OUCHI SHINKO KAGAKU

KOGYO Co., Ltd.)

\*6: Sodium 1,6-hexamethylenedithiosulfate dihydrate

\*7: KAYARAD D310 (a trade mark; manufactured by NIPPON KAYAKU Co., Ltd.)

\*8: Range of the change in storage modulus in the temperature range of 170 to 200°C

As shown in Table 1, the rubber composition of the present invention comprising the agent for preventing heat aging had, in the curve exhibiting the change in dynamic storage modulus during elevation of temperature, the intersection of the extrapolation line A of the portion in which the dynamic storage modulus shows an approximately linear change before rapidly decreasing at temperatures higher than 100°C and the extrapolation line B of the portion in which the dynamic storage modulus rapidly decreases, at a temperature of 170°C or higher independently of the type of the rubber component and the amount of carbon black. In Examples 12 to 16 in which the agent for preventing degradation was used in combination, the change in the dynamic storage modulus with temperature in the temperature range of 170 to 200°C decreased.

Radial tires having a size 225/60R16 were prepared in accordance with a conventional process using the above rubber compositions as the rubber composition for the rubber reinforcing layers disposed at the side wall portions and were subjected to the durability test. The results are shown in Table 1. In Examples 17 and 18 shown in Table 1, the same rubber compositions were used for the rubber reinforcing layers and for the bead fillers.

As shown by the results in Table 1, durability in the run-flat

condition can be improved by using the rubber composition of the present invention as the rubber composition for the rubber reinforcing layers. As shown by the results of Examples 17 and 18, durability of the tire in the run-flat durability can be further improved by using the rubber composition of the present invention as the rubber compositions for the rubber reinforcing layers and for the bead fillers.

Examples 19-24:

Rubber compositions were prepared in accordance with the formulations shown in Table 2 and the dynamic storage modulus  $E'$  was measured with an elevation of temperature. The results are shown in Table 2.

Table 2

Example		19	20	21	22	23	24
Comparative Example	2						
Natural rubber	30.0	30.0	30.0	30.0	30.0	30.0	30.0
Butadiene rubber * <sup>1</sup>	70.0	70.0	70.0	70.0	70.0	70.0	70.0
Carbon black * <sup>2</sup>	60.0	60.0	60.0	60.0	60.0	60.0	60.0
Softener * <sup>3</sup>	5.0	5.0	5.0	5.0	5.0	5.0	5.0
Zinc oxide	3.0	3.0	3.0	3.0	3.0	3.0	3.0
Stearic acid	1.0	1.0	1.0	1.0	1.0	1.0	1.0
Antioxidant * <sup>4</sup>	2.0	2.0	2.0	2.0	2.0	2.0	2.0
Vulcanization accelerator * <sup>5</sup>	3.5	3.5	3.5	3.5	3.5	3.5	3.5
Agent for preventing heat aging * <sup>6</sup>	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Agent for preventing degradation A * <sup>7</sup>	0.0	3.0	5.0	10.0	0.0	0.0	0.0
Agent for preventing degradation B * <sup>8</sup>	0.0	0.0	0.0	0.0	3.0	5.0	10.0
Sulfur	5.0	5.0	5.0	5.0	5.0	5.0	5.0
$\Delta E'$ * <sup>9</sup> (MPa)	2.5	1.8	1.1	0.7	2.3	1.8	1.3
Applied portion							
reinforcing rubber	applied	applied	applied	applied	applied	applied	applied
bead filler	not applied	not applied	not applied	not applied	not applied	not applied	not applied
Durability in the run-flat condition	100	108	125	151	102	108	118

The amounts in the above table are all in parts by weight.

\*1: BR01 (a trade mark; manufactured by JSR Corporation)

\*2: FEF

\*3: Spindle oil

\*4: NOCRAC 6C (a trade mark; manufactured by OUCHI SHINKO KAGAKU KOGYO Co., Ltd.)

\*5: NOCCELOR NS (a trade mark; manufactured by OUCHI SHINKO KAGAKU  
KOGYO Co., Ltd.)

\*6: Sodium 1,6-hexamethylenedithiosulfate dihydrate

\*7: TMPTA (a trade mark; manufactured by NIPPON KAYAKU Co., Ltd.)

\*8: KAYARAD D310 (a trade mark; manufactured by NIPPON KAYAKU Co., Ltd.)

\*9: Difference between max. and min. of storage modulus at a temperature between 180  
and 200°C

As shown in Table 2, the rubber composition of the present invention comprising the agent for preventing degradation had, in the curve exhibiting the change in dynamic storage modulus during elevation of temperature, a difference  $\Delta E'$  of 2.5 MPa or less ( $\Delta E'$ : a difference between the maximum value and the minimum value of the dynamic storage modulus at a temperature between 180 and 200°C).

Radial tires having a size 225/60R16 were prepared in accordance with a conventional process using the above rubber compositions as the rubber composition for the rubber reinforcing layers disposed at the side wall portions and were subjected to the durability test. The results are shown in Table 2.

WHAT IS CLAIMED IS:

1. A rubber composition having, in a curve exhibiting a change in dynamic storage modulus during elevation of temperature, an intersection of an extrapolation line A of a portion in which the dynamic storage modulus shows an approximately linear change before a rapid decrease at temperatures higher than 100°C and an extrapolation line B of a portion in which the dynamic storage modulus rapidly decreases, at a temperature of 170°C or higher.
2. A rubber composition according to claim 1, comprising sodium 1,6-hexamethylenedithiosulfate dihydrate.
3. A rubber composition according to claim 2, wherein the amount of sodium 1,6-hexamethylenedithiosulfate dihydrate is 1 to 10 parts by weight per 100 parts by weight of a rubber component.
4. A rubber composition according to claim 1, comprising a compound A having two or more ester groups in one molecule.
5. A rubber composition according to claim 4, wherein the compound A is an acrylate or a methacrylate.
6. A rubber composition according to claim 4, wherein the compound A is a polyfunctional ester of a polyhydric alcohol and acrylic acid or methacrylic acid, and wherein the polyhydric alcohol is at least one compound selected from the group consisting of tetramethylolmethane, trimethylolpropane and polymers of these compounds.
7. A rubber composition according to claim 6, wherein the polyhydric alcohol is trimethylolpropane or a dimer of

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tetramethylolmethane.

8. A rubber composition according to claim 4, wherein the amount of the compound A is 0.5 to 20 parts by weight per 100 parts by weight of a rubber component.

✓ 9. A rubber composition having, in a curve exhibiting a change in dynamic storage modulus during elevation of temperature, a difference  $\Delta E'$  between the maximum value and the minimum value of the dynamic storage modulus at a temperature between 180 and 200°C of 2.5 MPa or less.

10. A rubber composition according to claim 9, comprising a compound A having two or more ester groups in one molecule.

11. A rubber composition according to claim 10, wherein the compound A is an acrylate or a methacrylate.

12. A rubber composition according to claim 10, wherein the compound A is a polyfunctional ester of a polyhydric alcohol and acrylic acid or methacrylic acid.

13. A rubber composition according to claim 12, wherein the polyhydric alcohol is at least one compound selected from the group consisting of tetramethylolmethane, trimethylolpropane and polymers of these compounds.

14. A rubber composition according to claim 12, wherein the polyhydric alcohol is trimethylolpropane or a dimer of tetramethylolmethane.

15. A rubber composition according to claims 10, wherein the amount of the compound A is 0.5 to 20 parts by weight per 100 parts by weight of a rubber component.

16. A pneumatic tire comprising bead fillers and/or side wall portions reinforced with a rubber reinforcing layer, wherein a rubber composition for the bead fillers and/or the rubber reinforcing layer comprises the rubber composition according to claim 1.

17. A pneumatic tire comprising bead fillers and/or side wall portions reinforced with a rubber reinforcing layer, wherein a rubber composition for the bead fillers and/or the rubber reinforcing layer comprises the rubber composition according to claim 9.

18. A pneumatic tire according to claim 1, which is a run-flat tire.

✓19. A rubber composition for side reinforcing layers and/or bead fillers comprising sodium 1,6-hexamethylenedithiosulfate dihydrate.

20. A rubber composition according to claim 19, comprising a compound A having two or more ester groups in one molecule.



## ABSTRACT OF THE DISCLOSURE

A rubber composition having excellent heat resistance and a pneumatic tire produced by using this rubber composition are provided.

The rubber composition has, in a curve exhibiting a change in dynamic storage modulus during elevation of temperature, an intersection of an extrapolation line of a portion in which the dynamic storage modulus is approximately constant before rapidly decreasing at temperatures higher than 100°C and an extrapolation line of a portion in which the dynamic storage modulus rapidly decreases, at a temperature of 170°C or higher.

FIG. 1

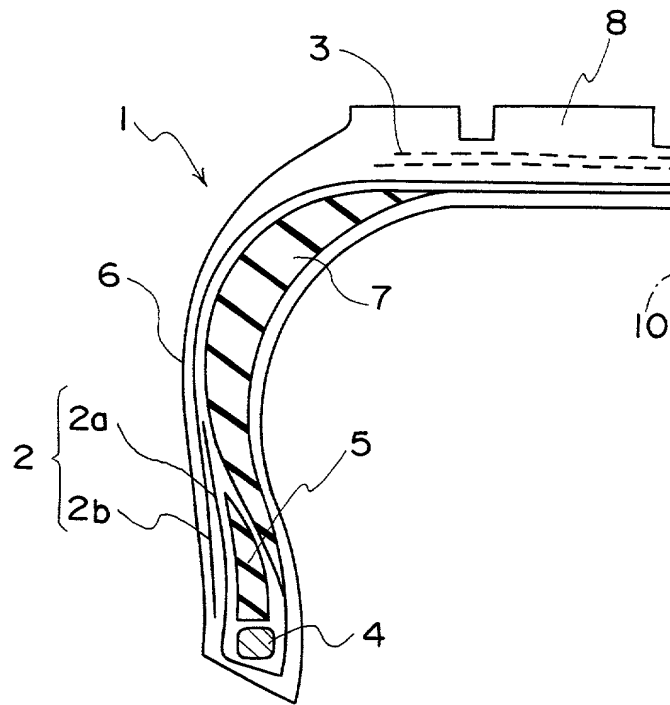


FIG. 2

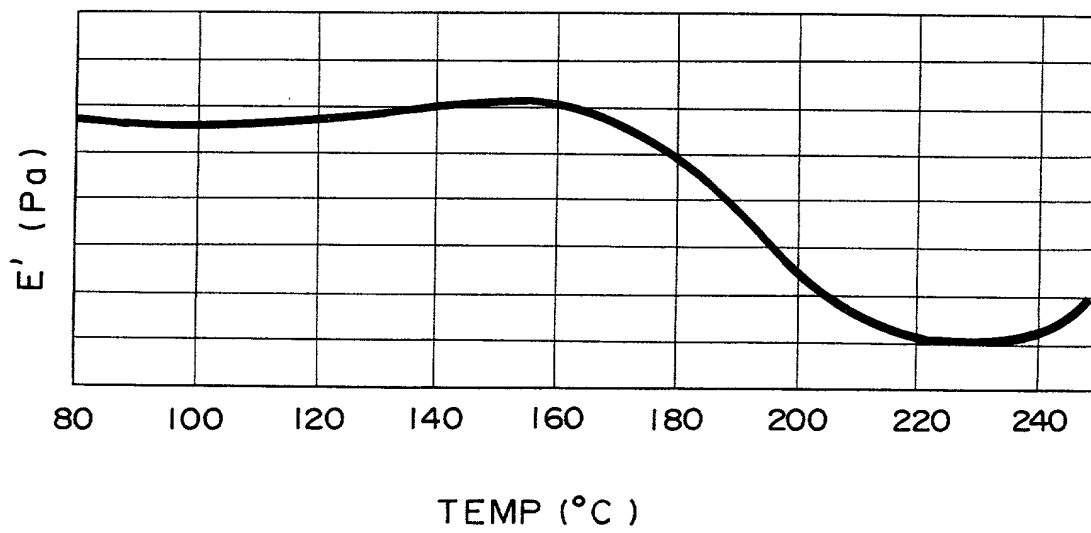
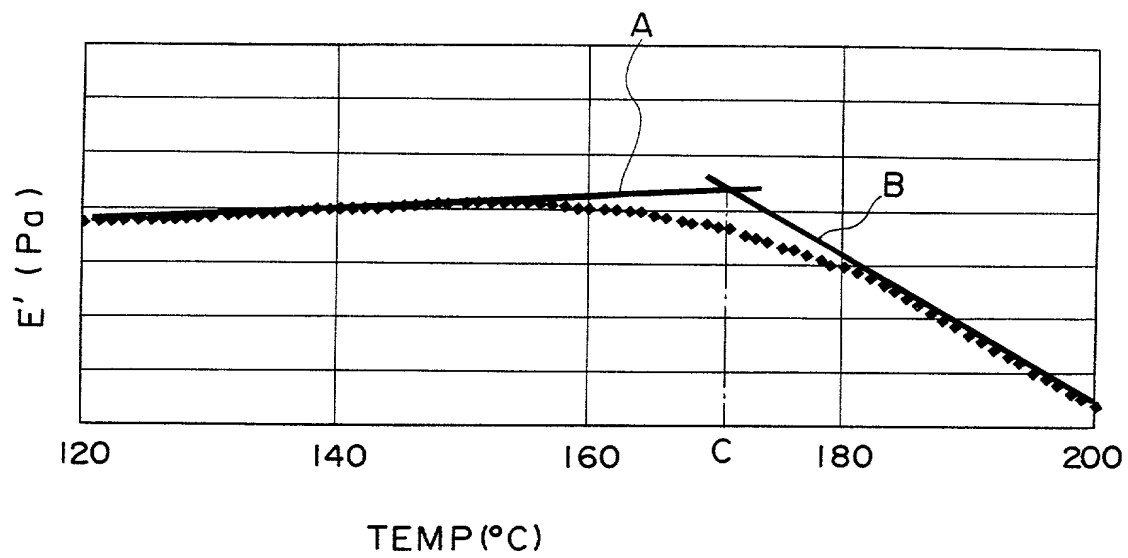


FIG. 3



# Declaration and Power of Attorney for Patent Application

特許出願宣言書及び委任状

## Japanese Language Declaration

日本語宣言書

下記の氏名の発明者として、私は以下の通り宣言します。

As a below named inventor, I hereby declare that:

私の住所、私書箱、国籍は下記の私の氏名の後に記載された通りです。

My residence, post office address and citizenship are as stated next to my name,

下記の名称の発明に関して請求範囲に記載され、特許出願している発明内容について、私が最初かつ唯一の発明者（下記の氏名が一つの場合）もしくは最初かつ共同発明者であると（下記の名称が複数の場合）信じています。

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

RUBBER COMPOSITION AND PNEUMATIC

TIRE USING SAID RUBBER COMPOSITION

上記発明の明細書（下記の欄でX印がついていない場合は、本書に添付）は、

the specification of which is attached hereto unless the following box is checked:

☐ \_\_\_\_月 \_\_\_\_日に提出され、米国出願番号または特許協定条約

国際出願番号を \_\_\_\_\_ とし、

（該当する場合） \_\_\_\_\_ に訂正されました。

☐ was filed on \_\_\_\_\_ as United States Application Number or PCT International Application Number

\_\_\_\_\_ and was amended on

\_\_\_\_\_ (if applicable).

私は、特許請求範囲を含む上記訂正後の明細書を検討し、内容を理解していることをここに表明します。

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

私は、連邦規則法典第37編第1条56項に定義されるとおり、特許資格の有無について重要な情報を開示する義務があることを認めます。

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56.

# Japanese Language Declaration

(日本語宣言書)

私は、米国法典第35編第119条(a)-(d)項又は第365条(b)項に基づき下記の、米国以外の国の少なくとも一カ国を指定している特許協力条約第365条(a)項に基づく国際出願、又は外国での特許出願もしくは発明者証の出願についての外国優先権をここに主張するとともに、優先権を主張している本出願の前に出願された特許または発明者証の外国出願を以下に、枠内をマークすることで、示しています。

I hereby claim foreign priority under Title 35, United States Code, Section 119(a)-(d) or 365(b) of any foreign application(s) for patent or inventor's certificate, or 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed.

## Prior Foreign Applications

外国での先行出願

## Priority Not Claimed

優先権主張なし

10-159512  
(Number)  
(番号)

Japan  
(Country)  
(国名)

8/June/1998  
(Day/Month/Year Filed)  
(出願年月日)

☐

(Number)  
(番号)

(Country)  
(国名)

(Day/Month/Year Filed)  
(出願年月日)

☐

(Number)  
(番号)

(Country)  
(国名)

(Day/Month/Year Filed)  
(出願年月日)

☐

私は、下記の米国法典第35編第120条に基づいて下記の米国特許出願に記載された権利、又は米国を指定している特許協力条約第365条(c)に基づく権利をここに主張します。又、本出願の各請求範囲の内容が米国法典第35編第112条第1項又は特許協力条約で規定された方法で先行する米国特許出願に開示されていない限り、その先行米国出願書提出日以降で本出願書の日本国内又は特許協力条約国際出願提出日までの期間中に入手された、連邦規則法典第37編第1条第56項で定義された特許資格の有無に関する重要な情報について開示義務があることを認識しています。

I hereby claim the benefit of Title 35, United States Code Section 120 of any United States application(s), or 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of Title 35, United States Code Section 112, I acknowledge the duty to disclose any material information which is material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application:

(Application No.)  
(出願番号)

(Filing Date)  
(出願日)

(Status: Patented, Pending, Abandoned)  
(現況: 特許許可済、係属中、放棄済)

(Application No.)  
(出願番号)

(Filing Date)  
(出願日)

(Status: Patented, Pending, Abandoned)  
(現況: 特許許可済、係属中、放棄済)

私は、私自身の知識に基づいて本宣言中で私が行う表明が真実であり、かつ私の入手した情報と私の信ずるところに基づく表明が全て真実であると信じていること、さらに故意になされた虚偽の表明及びそれと同等の行為は米国法典第18編第1001条に基づき、罰金または拘禁、もしくはその両方により処罰されること、そしてそのような故意による虚偽の声明を行えば、出願した、又は既に許可された特許の有効性が失われることを認識し、よってここに上記のごとく宣誓を致します。

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

# Japanese Language Declaration

(日本語宣言書)

委任状：私は、下記の発明者として、本出願に関する一切の  
手続を米国特許商標局に対して遂行する弁理士又は代理人  
として、下記のことを指名致します。(弁護士、又は代理人の  
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# Japanese Language Declaration

(日本語宣言書)

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国籍		Citizenship	
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住所		Residence	
国籍		Citizenship	
郵便の宛先		Post office address	